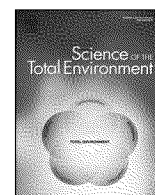




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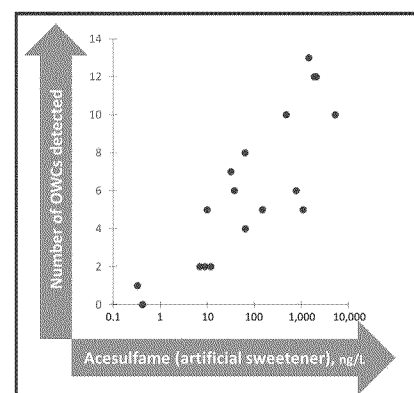
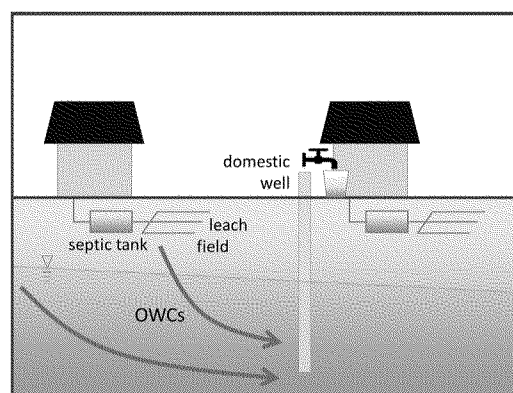
## Septic systems as sources of organic wastewater compounds in domestic drinking water wells in a shallow sand and gravel aquifer

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### HIGHLIGHTS

- We tested 20 domestic drinking water wells for 117 organic wastewater compounds.
- PFASs, pharmaceuticals, and an artificial sweetener were most frequently detected.
- Nitrate, boron, and well depth were all correlated with PFASs and pharmaceuticals.
- Acesulfame (artificial sweetener) is a sensitive marker of OWCs in groundwater.
- Septic systems are likely the main source; landfills may also affect some wells.

### GRAPHICAL ABSTRACT



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### abstract

Domestic drinking water wells serve 44 million people in the US and are common globally. They are often located in areas served by onsite wastewater treatment systems, including septic systems, which can be sources of biological and chemical pollutants to groundwater. In this study we tested 20 domestic drinking water wells in a sand and gravel aquifer on Cape Cod, Massachusetts, USA, for 117 organic wastewater compounds (OWCs) and for inorganic markers of septic system impact. We detected 27 OWCs, including 12 pharmaceuticals, five per- and polyfluoroalkyl substances (PFASs), four organophosphate flame retardants, and an artificial sweetener (acesulfame). Maximum concentrations of several PFASs and pharmaceuticals were relatively high compared to public drinking water supplies in the US. The number of detected OWCs and total concentrations of pharmaceuticals and of PFASs were positively correlated with nitrate, boron, and acesulfame and negatively correlated with well depth. These wells were all located in areas served exclusively by onsite wastewater treatment systems, which are likely the main source of the OWCs in these wells, although landfill leachate may also be a source. Our results suggest that current regulations to protect domestic wells from pathogens in septic system discharges do not prevent OWCs from reaching domestic wells, and that nitrate, a commonly measured drinking water contaminant, is a useful screening tool for OWCs in domestic wells. Nitrate concentrations of 1 mg/L  $\text{NO}_3\text{-N}$ , which are tenfold higher than local background and tenfold lower than the US federal drinking water standard, were associated with wastewater impacts from OWCs in this study.

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## 1. Introduction

Domestic drinking water wells, which serve 14% of the US population (44 million residents; Maupin et al., 2014) and are common globally, are often impacted by wastewater and other contamination sources. They are often shallower than large volume public supply wells and thus more vulnerable to contaminants, which often exceed drinking water standards. Unlike public supply wells, domestic wells are not regulated under the US EPA's Safe Drinking Water Act and many well owners do not regularly test their well water quality. In 2009, the US Centers for Disease Control and Prevention initiated its Unregulated Drinking Water Initiative to address domestic well water quality. One of the Initiative's goals was to better understand current water quality conditions in domestic wells throughout the US (Backer and Tosta, 2011). In a sampling of over 3800 domestic wells in Wisconsin, USA, 47% exceeded at least one health guideline for nitrate, coliform bacteria, fluoride, or metals (Knobloch et al., 2013), and a compilation of domestic well testing throughout the US showed exceedances of drinking water standards in 8.4% of wells for nitrate and 11% of wells for arsenic (Focazio et al., 2006).

Domestic wells are commonly used in communities that are served by onsite wastewater treatment systems (e.g., septic systems, cess-pools), which can be sources of pathogens and chemical contaminants to groundwater. Leachate from septic systems is likely to contaminate domestic wells in areas with high septic system density (Bremer and Harter, 2012). Coliform bacteria counts and nitrate and phosphate concentrations were higher in domestic wells closer to septic tanks in Florida (Arnade, 1999), and diarrheal disease in children was associated with density of nearby septic systems in Wisconsin (Borchardt et al., 2003).

In addition to conventional pollutants like nitrate and coliform bacteria, septic systems are also sources of organic wastewater compounds (OWCs), such as pharmaceuticals, personal care products, and organo-phosphate flame retardants (Hinkle et al., 2005; Swartz et al., 2006; Conn et al., 2010). Some OWCs are endocrine disrupting compounds (EDCs) that can alter hormone signaling; some have been linked to reproductive effects in fish and other freshwater organisms (Brian et al., 2007) and recent studies have suggested a growing number of human health endpoints associated with EDC exposure (WHO/UNEP, 2013). Incomplete degradation or sorption during treatment in septic tanks and leach fields, as well as leaks of poorly treated sewage from aging and failing systems, allow some OWCs to percolate through vadose zone soils and enter groundwater. Some OWCs can persist during subsurface transport and end up in groundwater (Swartz et al., 2006; Phillipset al., 2015), surface water (Standley et al., 2008; Dougherty et al., 2010) and drinking water (Verstraeten et al., 2005; Schaider et al., 2014). We previously found 18 OWCs in public supply wells on Cape Cod, Massachusetts, a region served by a sand and gravel aquifer where 85% of residents rely on onsite wastewater treatment systems (Schaider et al., 2014). Verstraeten et al. (2005) detected 14 pharmaceuticals in domestic wells in a shallow sand and gravel aquifer in central US and Erickson et al. (2014) detected several pharmaceuticals in domestic wells in both glacial and bedrock aquifers. However, despite widespread reliance on domestic wells and their vulnerability to pollution from septic systems and other sources, there is little information about the types and concentrations of OWCs in domestic wells.

The goals of this study were: (1) to measure OWC concentrations in domestic wells in areas served exclusively by onsite wastewater treatment systems; (2) to compare these concentrations with reports on other drinking water sources; and (3) to evaluate whether the presence of OWCs in domestic wells is correlated with other factors that may be proxies for septic system impact. Understanding the types and concentrations of OWCs in drinking water provides a basis for assessing OWC exposure and health risks from consumption of drinking water contaminated by household wastewater. Our results also provide insight into the characteristics of drinking water wells most likely to contain

OWCs, which can inform source water protection and drinking water quality monitoring.

## 2. Materials and methods

### 2.1. Selection of wells

To select wells for OWC analysis with a wide range of septic system impact, we recruited participants throughout Cape Cod (Barnstable County), Massachusetts, USA and used a combination of GIS land use analysis and nitrate and boron testing to select 20 homes for sampling. Eighty-five percent of Cape Cod residents are served by onsite wastewater treatment systems and 20% rely on domestic drinking water wells. We recruited participants through electronic mailings, posters in public buildings, and coverage in local media.

For each of 110 wells whose owners volunteered for the study and provided an address, we used ArcMap (ESRI, Redlands, CA) to analyze land use within a capture zone around each well. To identify capture zones, we followed Kerfoot and Horsley's (1988) methodology for developing protective zones around domestic wells on Cape Cod. This method incorporates typical groundwater velocities and accounts for potential seasonal fluctuations in the direction of groundwater flow and pumping rates on Cape Cod. The shape of each capture zone is roughly elliptical, with a 30-m radius drawn around a line that starts at the well and extends 60 m in an upgradient direction (total area: 6500 m<sup>2</sup>). Within each capture zone, we calculated the fraction of the area used for varying densities of residential development. We used 2005 land cover/land use data from MassGIS (Massachusetts Information Technology Division, 2012), which included 33 land use types and had a 0.5 m resolution. We used the results of the land use analysis to calculate an average number of homes per unit area (average density) within each protective zone and to calculate the total fraction of land area within each capture zone used for residential development (%RES). Additional information about the process is provided in Supplementary Material. Of the 110 candidate wells, we selected 50 wells as follows: 20 wells with the highest average density (1.2–4.5 homes/acre), 15 wells with the lowest average density (b0.14 homes/acre), and 15 wells with intermediate average density (0.2–1 homes/acre). In selecting wells with the lowest and highest average density, we excluded several wells because they were within 400 m of another well with similar density, and in selecting wells with intermediate density, we prioritized several towns (Eastham, Wellfleet, Truro) that rely almost exclusively on domestic wells, while also aiming to include wells throughout Cape Cod.

In order to gain a more accurate assessment of potential septic system impact in each well, we asked each of the 50 selected households to collect a water sample for nitrate (NO<sub>3</sub><sup>-</sup>) and boron (B) analysis and to complete a questionnaire about well depth, results of prior water quality monitoring, and known water quality concerns. We used these results to select 20 wells for OWC analysis. While our well selection was not designed to be a statistical representation of domestic wells across Cape Cod, we aimed to include wells with low, medium, and high NO<sub>3</sub><sup>-</sup> concentrations and a wide range of residential land use density in their capture zones. Of the 43 wells that we tested for NO<sub>3</sub><sup>-</sup> and B, we selected 20 wells for OWC analysis as follows, using categories developed for Cape Cod groundwater (Massachusetts EOE, 2004): 5 of 14 wells with low NO<sub>3</sub><sup>-</sup> (b0.5 mg/L) and B (≤20 µg/L), 6 of 13 wells with moderate NO<sub>3</sub><sup>-</sup> (0.5–2.5 mg/L) and B (20–50 µg/L), and all 9 wells with high nitrate (≥2.5 mg/L). We oversampled wells with NO<sub>3</sub><sup>-</sup> above 2.5 mg/L (45% of final 20 wells compared to 21% of the 43 wells tested for NO<sub>3</sub><sup>-</sup> and B) to more thoroughly characterize OWC concentrations in wells likely to be most impacted. Within the low and moderate NO<sub>3</sub><sup>-</sup> categories, we selected wells with a wide range of residential land use density (b0.1–2.3 homes/acre). Concentrations of NO<sub>3</sub><sup>-</sup> and B presented in this paper are from the second round of sampling collected at the same time as samples analyzed for OWCs.

## 2.2. Sample collection for OWC analysis

Raw water samples were collected from all 20 wells by study team members in February 2011. To avoid possible sample contamination by target compounds, personnel refrained from consuming caffeinated beverages and using over-the-counter medications, antimicrobial products, sunscreens, and insect repellent on the day of sampling. In homes with whole-house water treatment (e.g., pH adjustment, filtration for iron and manganese), we collected samples from spigots prior to the water treatment system. In homes with no whole-house treatment, samples were collected from sinks and bathtubs. We flushed each tap or spigot for at least 10 min to pull fresh water from the ground.

## 2.3. Chemical analyses

We selected 117 target OWCs based on previous detections in aquatic systems, especially groundwater and drinking water, evidence of endocrine disruption, and/or availability of an analytical method. Ninety-two OWCs were also target analytes in our study of public supply wells on Cape Cod (Schaider et al., 2014). Method detection limits (MDLs) for OWC analyses varied by four orders of magnitude, from 0.02 to 280 ng/L, but most (74%) were below 10 ng/L and the majority (52%) were below 1 ng/L. A complete list of 117 target OWC analytes and MDLs is provided in Table S1.

Details of analytical methods and data validation are provided in Supplementary material. Underwriters Laboratories (now Eurofins Eaton Analytical, Indiana, USA) conducted the analyses for OWCs,  $\text{NO}_3^-$ , B, and sodium. For OWC analysis, samples were spiked with isotopically labeled surrogate compounds and extracted with a solid phase extraction (SPE) cartridge. Extracts were spiked with internal standards to account for variations in the instrument signal when calculating recoveries of the surrogate standards. Pharmaceuticals and personal care products (PPCPs), hormones, per- and polyfluoroalkyl substances (PFASs), alkylphenols, and herbicides were analyzed by liquid chromatography/tandem mass spectrometry (LC–MS/MS). Organophosphate flame retardants were analyzed by large-volume injection gas chromatography/mass spectrometry (LVI GC/MS). For hormones and some PPCPs and PFASs with available isotopically labeled analogs, analytes were quantified using isotope dilution. For analytes without matched analogs, surrogate standard recoveries were compared to ranges of acceptable values but concentrations were not corrected for surrogate recoveries.

Inorganic constituents were measured by ion chromatography ( $\text{NO}_3^-$ ), inductively coupled plasma (ICP) mass spectrometry (B), and ICP atomic emission spectrometry (sodium). In this paper, nitrate concentrations are reported in units of  $\text{NO}_3\text{-N}$  (mg/L). Total nitrogen (TN) and total organic carbon (TOC) were analyzed using combustion methods by the Barnstable County Water Quality Laboratory (Massachusetts, USA).

## 2.4. Data analysis

We calculated correlations between pairs of individual PFASs and between metrics of septic system impact (e.g., concentrations of  $\text{NO}_3^-$  and B, well depth) and OWC presence using the nonparametric Spearman's rank correlation test. Correlations were considered significant for  $p \leq 0.05$ .

## 3. Results and discussion

### 3.1. Inorganic contaminants

Among the 20 wells that we tested for OWCs, concentrations of nitrate ( $\text{NO}_3^-$ ) varied by more than a factor of 100, and total nitrogen (TN) and boron (B) concentrations ranged by more than a factor of 25 (Table 1). Five wells had  $[\text{NO}_3^-]$  consistent with background or minimal

anthropogenic impact ( $\leq 0.5$  mg/L), five wells showed moderate impact ( $0.5\text{--}2.5$  mg/L), and 10 were highly impacted ( $\geq 2.5$  mg/L). One well was above USEPA's Maximum Contaminant Level (MCL) of 10 mg/L. On average, 89% of TN was present as  $\text{NO}_3^-$  (range: 75–96%), an indication of oxic conditions in all groundwater samples and minimal contributions from ammonia ( $\text{NH}_4^+$ ), organic N, or other N species. By comparison, in a study of 26 shallow wells in Nebraska, USA, the median  $[\text{NO}_3^-]$  was  $\leq 0.05$  mg/L and the maximum was 39 mg/L (Verstraeten et al., 2005). In Nebraska wells  $\leq 15$  m from septic systems, concentrations of  $\text{NH}_4^+$  exceeded those of  $\text{NO}_3^-$ , indicating reducing conditions. This close proximity between wells and septic systems may lead to insufficient residence times in the vadose zone for oxidation of  $\text{NH}_4^+$  to  $\text{NO}_3^-$  in septic system plumes. Among 3465 domestic wells tested throughout the US (Focazio et al., 2006), 90% had  $\leq 0.01$  mg/L  $\text{NO}_3^-$  (compared with 80% in our study), 8% had  $\geq 10$  mg/L  $\text{NO}_3^-$  (compared with 5% in our study), and the mean concentration was 3.6 mg/L (compared with 2.9 mg/L in our study). This comparison suggests that the distribution of  $[\text{NO}_3^-]$  in our study was similar to that of Focazio et al.'s broader nationwide study, although their sampling was not part of a nationally representative sampling design.

Boron (B) concentrations ranged from 6.3 to 250  $\mu\text{g/L}$ . The four wells with non-detectable  $\text{NO}_3^-$  had  $\leq 13$   $\mu\text{g/L}$  B, consistent with background concentrations in Cape Cod groundwater ( $\sim 10$   $\mu\text{g/L}$  B; Schaider et al., 2014). The two wells with the highest [B] (250 and 91  $\mu\text{g/L}$ ) also had two of the three highest  $[\text{NO}_3^-]$  ( $\geq 5$  mg/L), consistent with septic systems as the main source.

### 3.2. Occurrence of OWCs

Of the 117 OWCs that we analyzed, 27 compounds (23%) were detected in at least one well (Table 2, Fig. 1). Seventeen wells (85%) contained detectable concentrations of at least one OWC, with a maximum of 13 OWCs detected in a single well. Seven OWCs were detected in at least one-quarter of wells tested: an artificial sweetener (acesulfame), four per- and polyfluoroalkyl substances (PFASs), and two pharmaceuticals (sulfamethoxazole, carbamazepine). We also infrequently detected five organophosphate flame retardants and plasticizers and two hormones. Seventy-seven percent of OWCs were not detected in any sample, including all tested alkylphenols and herbicides. Table S1 provides a complete list of the 117 analytes and Table S3 provides concentrations in individual wells for all 27 detected compounds.

#### 3.2.1. Acesulfame

Acesulfame (or Ace K), an artificial sweetener, was the most frequently detected OWC, found in 17 wells (85%). It was detected in all samples with detectable concentrations of other OWCs. Concentrations ranged four orders of magnitude, from  $\leq 0.42$  up to 5300 ng/L. Our maximum concentration is higher than those reported in two other studies of raw drinking water from groundwater (2600 ng/L; Buerge et al., 2009) and surface water (4200 ng/L; Scheurer et al., 2010) sources (Table 2) and in a study of treated tap water (1600 ng/L; Spoelstra et al., 2013).

Table 1  
Characteristics of 20 domestic wells selected for OWC analysis.

	Mean <sup>a</sup>	Median	Minimum	Maximum	# non-detects
$\text{NO}_3\text{-N}$ (mg/L)	2.9	2.3	$\leq 0.1$	11	4
Total N (mg/L)	2.8	2.5	$\leq 0.5$	13	4
Boron ( $\mu\text{g/L}$ )	40	24	6.3	250	0
TOC (mg/L)	—	$\leq 1$	$\leq 1$	6.4	15
Sodium (mg/L)	22	20	8.6	45	0
Well depth (m) <sup>b</sup>	18	18	3	40	—

<sup>a</sup> Calculated mean concentrations of  $\text{NO}_3\text{-N}$  and total N were the same regardless of substitution method (either zero or MDL) for concentrations below the MDL.

<sup>b</sup>  $n = 18$ .

Table 2  
OWC concentrations detected in raw water samples from 20 domestic wells on Cape Cod, Massachusetts.

Chemical name	Method detection limit (MDL) (ng/L)	Number of times detected (%)	Maximum concentration (ng/L)	Health-based guideline values (ng/L)	Maximum concentrations reported in raw drinking water sources (ng/L)	
					Ground water	Surface water
Pharmaceuticals—antibiotics						
Monensin (Mon)	0.52	1 (5%)	0.8J		b1 <sup>q</sup>	1.4 <sup>j</sup> 2.4 <sup>u, +</sup>
Sulfachloropyridazine (SulfCP)	0.58	2 (10%)	0.7J		b5 <sup>f</sup> b5 <sup>q</sup>	b5 <sup>i</sup> b50 <sup>j</sup> 1.3 <sup>u</sup>
Sulfamethoxazole (SulfMX)	0.1	9 (45%)	60	440,000 <sup>s</sup> 18,000,000 <sup>d</sup>	41 <sup>g</sup> 58 <sup>x</sup> 82 <sup>f</sup> 113 <sup>g</sup> 150 <sup>w</sup> 170 <sup>h</sup>	b23 <sup>g</sup> 2 <sup>u</sup> 12 <sup>j</sup> 60 <sup>i</sup> 110 <sup>b</sup>
Sulfathiazole (SulfTZ)	0.27	1 (5%)	0.2J		b1 <sup>q</sup> b5 <sup>f</sup> b100 <sup>g</sup> b100 <sup>w</sup>	b50 <sup>j</sup> b100 <sup>g</sup> 0.4 <sup>u</sup>
Trimethoprim (Trim)	0.1	1 (5%)	1	6,700,000 <sup>d</sup>	b5 <sup>f</sup> b13 <sup>x</sup> 0.3 <sup>g</sup> 0.7 <sup>q</sup> 18 <sup>h</sup> 580 <sup>w</sup>	1 <sup>u</sup> 4 <sup>j</sup> 11 <sup>b</sup> 25 <sup>g</sup> 80 <sup>i</sup>
Pharmaceuticals—non-antibiotics						
Antipyrine (Antip)	0.83	1 (5%)	2		1 <sup>q</sup>	b1 <sup>j</sup>
Carbamazepine (CarbMP)	0.068	5 (25%)	62	1000 <sup>s</sup> 12,000 <sup>d</sup> 40,000 <sup>n</sup>	5.3 <sup>x</sup> 72 <sup>q</sup> 110 <sup>g</sup> 122 <sup>f</sup> 420 <sup>h</sup>	2 <sup>u</sup> 9 <sup>j</sup> 51 <sup>b</sup> 156 <sup>i</sup> 190 <sup>g</sup> 600 <sup>t</sup>
Cotinine (Cot)	0.59	1 (5%)	1		b1 <sup>q</sup> b14 <sup>x</sup> b19 <sup>h</sup> 60 <sup>w</sup> 102 <sup>g</sup>	10 <sup>i</sup> 12 <sup>j</sup> 74 <sup>g</sup>
Gemfibrozil (Gem)	0.15	1 (5%)	0.3 J	14,000 <sup>d</sup>	b13 <sup>x</sup> b15 <sup>g</sup> 1.2 <sup>q</sup>	b13 <sup>t</sup> b15 <sup>g</sup> 4 <sup>u</sup> 17 <sup>j</sup> 24 <sup>b</sup>
Meprobamate (Mep)	0.1	3 (15%)	2	260,000 <sup>e</sup>	5.4 <sup>q</sup>	73 <sup>b</sup>
Primidone (Prim)	2.1	2 (10%)	9			35 <sup>i</sup>
Simvastatin (Sim)	3	1 (5%)	14	4900 <sup>e</sup>	b5 <sup>q</sup>	b0.25 <sup>b</sup> b1 <sup>j</sup>
Per- and polyfluoroalkyl substances (PFASs)						
PFBS	0.22	11 (55%)	23	7000 <sup>k</sup>	3.7 <sup>a</sup> 6 <sup>o</sup> 9 <sup>c</sup>	1 <sup>u</sup> 5 <sup>c</sup> 6 <sup>o</sup> 47 <sup>a</sup>
PFHpA	0.25	6 (30%)	1J		33 <sup>a</sup> 39 <sup>c</sup> 54 <sup>o</sup>	b4 <sup>c</sup> 10 <sup>o</sup> 12 <sup>a</sup>
PFHxS	0.33	11 (55%)	41		9.3 <sup>p</sup> 10 <sup>o</sup> 11 <sup>a</sup> 32 <sup>c</sup>	8 <sup>c</sup> 12 <sup>p</sup> 18 <sup>a</sup> 46 <sup>o</sup>
PFHxA	0.16	10 (50%)	2		14 <sup>p</sup> 28 <sup>c</sup> 67 <sup>a</sup> 110 <sup>o</sup>	17 <sup>o</sup> 29 <sup>p</sup> 38 <sup>a</sup> 139 <sup>c</sup>
PFOS	0.24	11 (55%)	7	200 <sup>v</sup> 300 <sup>k</sup>	12 <sup>o</sup> 27 <sup>a</sup> 41 <sup>p</sup> 50 <sup>c</sup> 97 <sup>q</sup>	16 <sup>u</sup> 22 <sup>p</sup> 43 <sup>o</sup> 47 <sup>a</sup> 62 <sup>c</sup>
Flame retardants and plasticizers						
Bisphenol A (BPA)	2.5	1 (5%)	4J		b200 <sup>f</sup> b1000 <sup>x</sup> 450 <sup>g</sup>	14 <sup>b</sup> 360 <sup>t</sup> 1900 <sup>g</sup>

Table 2 (continued)

Chemical name	Method detection limit (MDL) (ng/L)	Number of times detected (%)	Maximum concentration (ng/L)	Health-based guideline values (ng/L)	Maximum concentrations reported in raw drinking water sources (ng/L)	
					Ground water	Surface water
2-EHDP	1.5	2 (10%)	18		b10 <sup>q</sup>	2000 <sup>u</sup>
TBP	5.1	1 (5%)	11		b10 <sup>q</sup> b160 <sup>f</sup> 190 <sup>x</sup> 420 <sup>g</sup>	140 <sup>i</sup> 740 <sup>g</sup>
TEP	10	1 (5%)	38	1,950,000 <sup>h</sup>	20 <sup>q</sup>	
TPP	1.5	1 (5%)	14		b10 <sup>q</sup> 46 <sup>x</sup> 67 <sup>g</sup>	b500 <sup>g</sup> 80 <sup>i</sup>
Hormones						
Cis-testosterone (Test)	0.029	1 (5%)	0.04J		b0.1 <sup>q</sup> b0.8 <sup>f,^</sup>	
Progesterone (Prog)	0.028	3 (15%)	0.04J		b0.1 <sup>q</sup> b8 <sup>f</sup>	0.5 <sup>u</sup> 3.1 <sup>b</sup>
Other						
Acesulfame (AceK)	0.42	17 (85%)	5300		2600 <sup>h</sup>	4200 <sup>r</sup>
N,N-diethyl-meta-toluamide (DEET)	0.67	3 (15%)	4J	200,000 <sup>m</sup> 6,250,000 <sup>s</sup>	6 <sup>q</sup> 74 <sup>x</sup> 410 <sup>g</sup>	16 <sup>i</sup> 110 <sup>b</sup> 200 <sup>i</sup> 270 <sup>f</sup>
Salicylic acid (Sal)	15	3 (15%)	30J			

References (number of samples tested in parentheses if provided. GW = groundwater, SW = surface water):

- <sup>a</sup> Appleman et al. (2014) (7 GW, 25 SW, 1 blend).  
<sup>b</sup> Benotti et al. (2009) (18 SW, 1 GW).  
<sup>c</sup> Boiteux et al. (2012) (196 GW, 135 SW).  
<sup>d</sup> Bruce et al. (2010).  
<sup>e</sup> Buerge et al. (2009) (GW).  
<sup>f</sup> Erickson et al. (2014) (29 GW, all domestic wells).  
<sup>g</sup> Focazio et al. (2008) (25 GW, 49 SW).  
<sup>h</sup> Fram and Belitz (2011) (1231 GW).  
<sup>i</sup> Guo and Krasner (2009) (7 SW).  
<sup>j</sup> Illinois EPA (2008) (5 SW).  
<sup>k</sup> Minnesota DOH (2008).  
<sup>m</sup> Minnesota Department of Health (2011a).  
<sup>n</sup> Minnesota Department of Health (2011b).  
<sup>o</sup> Post et al. (2013) (17 or 52 GW (varies by analyte), 13 SW).  
<sup>p</sup> Quiñones and Snyder (2009) (3 GW, 4 SW).  
<sup>q</sup> Schaider et al. (2014) (20 GW).  
<sup>r</sup> Scheurer et al. (2010) (6 SW).  
<sup>s</sup> Schriks et al. (2010).  
<sup>t</sup> Stackelberg et al. (2007) (12 SW, 1 location).  
<sup>u</sup> Tabe (2010) (22 SW, 2 locations).  
<sup>v</sup> US EPA (2009).  
<sup>w</sup> Verstraeten et al. (2005) (26 GW, all domestic wells).  
<sup>x</sup> Zimmerman (2005) (8 GW, 4 domestic wells).  
<sup>+</sup> Concentration reported for monensin sodium.  
<sup>^</sup> Isomer not specified for testosterone.

### 3.2.2. Pharmaceuticals

We detected twelve pharmaceuticals, including five antibiotics, out of a total of 59 pharmaceutical analytes, and 13 wells (65%) had detectable concentrations of at least one pharmaceutical. The two most frequently detected pharmaceuticals, which also had the highest maximum concentrations, were sulfamethoxazole (9 wells above 0.1 ng/L; maximum 60 ng/L) and carbamazepine (5 wells above 0.07 ng/L; maximum 62 ng/L). Sulfamethoxazole and carbamazepine have been among the most frequently detected OWCs in drinking water and groundwater (Barnes et al., 2008; Benotti et al., 2009; Erickson et al., 2014; Schaider et al., 2014). The maximum sulfamethoxazole concentration in this study matched or exceeded the maximum concentration in five drinking water studies (Zimmerman, 2005; Stackelberg et al., 2007; Focazio et al., 2008; Illinois EPA, 2008; Tabe, 2010), and was within a factor of 3 of the maximum concentration in five other studies (Verstraeten et al., 2005; Benotti et al., 2009; Fram and Belitz, 2011; Erickson et al., 2014; Schaider et al., 2014). Simvastatin was detected in one well (14 ng/L), the first reported detection in

drinking water. It was not detected in 20 public wells on Cape Cod (b5 ng/L, Schaider et al., 2014), in 19 US drinking water sources (b1 ng/L, Benotti et al., 2009), or in 5 drinking water sources in Illinois (b1 ng/L, Illinois EPA, 2008).

Nine pharmaceuticals were detected in only one or two wells, consistent with a high degree of heterogeneity in groundwater pharmaceutical concentrations. Heterogeneous concentrations are to be expected, particularly for prescription medications that are used by a small portion of the population at any given time. As domestic wells have much smaller recharge areas than public supply wells, the presence of prescription medications depended on whether the well pulled from an area where a resident was using that medication and the persistence of the compound in groundwater.

In a study of 26 domestic wells in Nebraska (Verstraeten et al., 2005), the maximum concentrations for seven of the pharmaceuticals we detected were higher than the maximum concentrations that we measured on Cape Cod by factors of 2.5 (sulfamethoxazole) to 580 (trimethoprim). The range of well depths was similar in both studies (5–

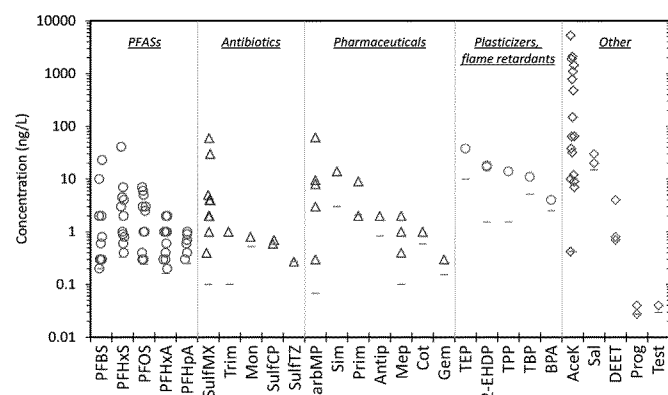


Fig. 1. OWC concentrations in 20 domestic wells on Cape Cod. Horizontal gray lines show method detection limits. Full chemical names provided in Table 2.

30 m in Nebraska, 3–40 m on Cape Cod). The relatively high concentrations in the Nebraska wells may be due to closer proximity between wells and septic systems or to older, less effective onsite treatment systems. Several OWCs detected in the Nebraska wells but not in the Cape Cod wells (acetaminophen, caffeine, paraxanthine) are well-removed in septic system leach fields (Wilcox et al., 2009; Katz et al., 2010) and typically only persist in anoxic groundwater (Swartz et al., 2006). Thus the presence of reducing conditions in the Nebraska aquifer likely promoted greater persistence of pharmaceuticals and other OWCs, whereas oxic conditions in the Cape Cod aquifer promoted greater microbial degradation.

### 3.2.3. PFASs

Four PFASs (PFOS, PFHxS, PFBS, and PFHxA) were detected in  $\geq 50\%$  of wells we tested, and another (PFHpA) was detected in 30% of wells. Six wells had detectable concentrations of all five of these PFASs, and one well had the highest concentrations of PFHxS (41 ng/L), PFBS (23 ng/L), and PFHxA (2 ng/L). PFOA was detected in laboratory method blanks (2.9, 3.2 ng/L) and field blanks (4 ng/L,  $n = 2$ ) at levels close to the maximum concentration in field samples (7 ng/L). While PFOA concentrations in most wells were  $\leq 4$  ng/L, five wells had PFOA concentrations of 2–3 ng/L after subtracting the field blank concentration; these five wells were among the six with detectable concentrations of five other PFASs.

We compared PFAS concentrations in domestic wells on Cape Cod to those reported in five other studies of PFASs in raw drinking water in the US and France (Quiñones and Snyder, 2009; Tabe, 2010; Boiteux et al., 2012; Post et al., 2013; Appleman et al., 2014). The maximum concentrations of two perfluoroalkanesulfonates, PFHxS and PFBS, on Cape Cod were generally higher than those reported in the other studies and were within a factor of two of the maximum concentrations reported in these studies (Table 2). Our maximum PFHxS concentration was also higher than 99.5% of treated tap water samples (NN 22,000) analyzed as part of US EPA's Unregulated Contaminant Monitoring Rule (UCMR3) testing (based on data presented in US EPA, 2015).

By contrast, maximum concentrations of PFOS and several perfluoroalkyl carboxylates in Cape Cod wells were substantially lower than those reported for raw water samples in the same five studies. The maximum concentrations of PFOS, PFHpA, and PFHxA that we measured were 14, 54, and 70 times lower, respectively, than those reported in the same five published studies. Our estimated maximum concentration of PFOA ( $\sim 3$  ng/L) is 40 times lower than the maximum reported concentration (120 ng/L; Appleman et al., 2014). We did not detect PFNA (b.0.45 ng/L) or PFDA (b.0.28 ng/L) in any of the samples tested on Cape Cod. While these two longer chain carboxylates are often less frequently detected than shorter-chain PFASs, maximum concentrations of PFNA (96 ng/L; Post et al., 2013) and PFDA (3.3 ng/L; Quiñones and Snyder, 2009) were much higher in other studies.

Overall, the relatively high concentrations of PFBS and PFHxS but not PFOS or  $C_{6-10}$  perfluoroalkyl carboxylates suggests that compared to other locations, the sources of PFASs into groundwater on Cape Cod are relatively enriched in the shorter-chain sulfonate forms, and that other types of sources (e.g., industrial) absent near domestic wells on Cape Cod may be relatively enriched in PFOS and perfluoroalkyl carboxylates.

Significant correlations were observed among the four most frequently detected PFASs, with Spearman rank correlation coefficients ( $\rho$ ) ranging from 0.61 to 0.86 ( $p < 0.01$  for all; Fig. 2). PFBS and PFHxS had the strongest correlation and most linear relationship, suggesting a common source, although temporal trends in production of these PFASs vary. Production of PFOS, PFHxS, and PFOA has declined in the US since the early 2000s following a phase-out of POSF-related chemicals by 3M and subsequent stewardship plans by US EPA, and this has resulted in declines in blood serum concentrations of these three PFASs in Americans (Calafat et al., 2007; Olsen et al., 2008). By contrast, PFBS is used as a replacement for POSF-related chemicals (e.g., PFOS and PFHxS) and PFHxA is used as a shorter-chain analog of PFOA (Wang et al., 2013).

### 3.2.4. Infrequently detected OWCs

Several classes of target analytes were detected in  $\leq 25\%$  of wells tested, including organophosphate flame retardants (OPFRs), hormones, and alkylphenols. Five wells (25%) had detectable concentrations of one of four non-halogenated OPFRs ranging from 11 to 38 ng/L. We did not detect chlorinated OPFRs, including three that we detected in public wells on Cape Cod (TECP, TDCPP, TCPP; Schaider et al., 2014), even though they tend to be more resistant to biodegradation than non-halogenated OPFRs (Meyer and Bester, 2004). The low detection frequencies for OPFRs in Cape Cod wells may reflect relatively low concentrations in domestic wastewater compared to wastewater in other locations with greater industrial, commercial, or institutional inputs. Phillips et al. (2015) reported 20  $\mu\text{g/L}$  TBEP in groundwater downgradient of a septic system serving an elder care facility where the most likely source was floor wax.

Among the nine hormone target analytes, only two were detected (progesterone and cis-testosterone), at relatively low concentrations (0.02–0.04 ng/L), in a total of four wells (20%). The low detection frequencies are likely due to biodegradation and sorption processes that limit hormone discharges from septic systems and transport in groundwater. Although we previously found three endogenous hormones in groundwater-fed ponds on Cape Cod (3–6.5 ng/L; Standley et al., 2008), we did not detect hormones in 20 public wells on Cape Cod (MRLs 0.1–0.5 ng/L; Schaider et al., 2014). Aerobics and filters in onsite wastewater treatment systems were shown to remove N90% of steroid estrogens (Stanford and Weinberg, 2010) and mass loss of 17 $\beta$ -estradiol was observed in a wastewater treatment plant plume on Cape Cod (Barber et al., 2009).

We did not detect nonylphenol (NP), octylphenol (OP), NP ethoxycarboxylates (NPECs), or NP ethoxylates (NPEOs) in the current study. These alkylphenols are metabolites of long-chain alkylphenol ethoxylates that are widely used as surfactants in detergents, personal care products, and plastics. One reason for the absence of detectable alkylphenols in our samples may be biotransformation in oxic groundwater. While NP and NPEOs have been measured at 10s to 100s of  $\mu\text{g/L}$  in septic tanks (Hinkle et al., 2005; Stanford and Weinberg, 2010), some studies have found N90% removal in septic system drainfields (Huntsman et al., 2006). NP, NPEOs, and NPECs were persistent in anoxic groundwater in a plume from a septic system leach pit but were not detected in oxic groundwater within the same plume (Swartz et al., 2006). The lack of detectable alkylphenols may be partially attributed to relatively high MDLs (10–92 ng/L) compared to MDLs of  $\sim 1$  ng/L for many other OWCs in our study (Table S1). The phase-out of nonylphenol ethoxylates from many laundry detergents (McCoy,



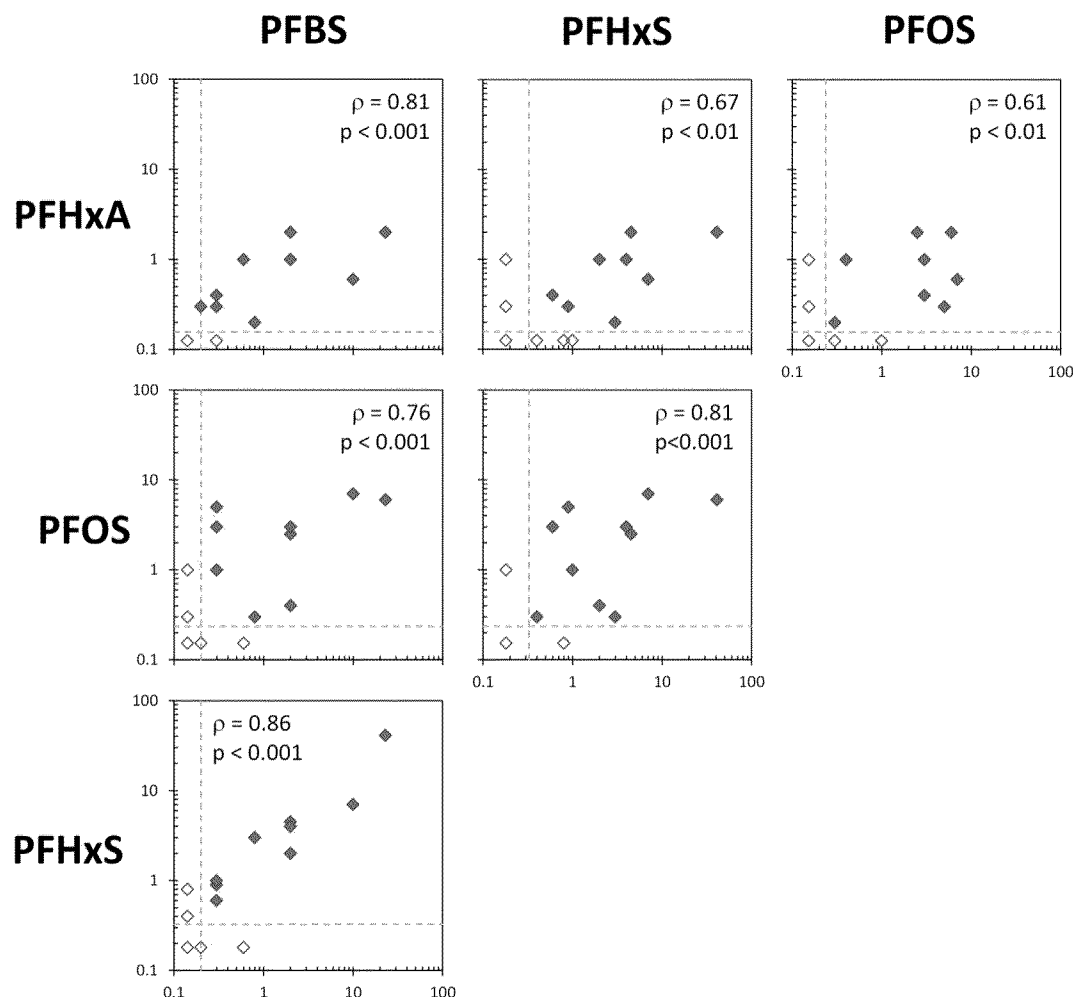


Fig. 2. Correlations between pairs of PFASs detected in at least 50% of domestic wells. Dashed gray lines show method detection limits (MDLs). Closed symbols show wells where both PFASs were NMDLs. Open symbols show wells with one or both PFASs bMDL, which were plotted between the MDL and axis as an approximation. For each pair, 6–8 wells had concentrations bMDL for both PFASs. Concentrations in ng/L.

2007) may also contribute to the lack of detections in the wells we tested.

### 3.3. Correlations between OWCs and indicators of septic system impact

We used three metrics of the presence of OWCs in domestic wells: total number of OWCs detected ( $N_{\text{detects}}$ ), sum of detected pharmaceutical concentrations ( $\Sigma[\text{pharma}]$ ), and sum of detected PFAS concentrations ( $\Sigma[\text{PFAS}]$ ).  $N_{\text{detects}}$  may include OWCs that originate from multiple pollution sources, whereas pharmaceuticals likely originate primarily from household wastewater. Septic systems may be a source of PFASs in groundwater because household wastewater can contain PFASs that have leached from food packaging, cookware, upholstery, apparel, and other household products. Landfill leachate, street runoff, and atmospheric deposition also can be sources of PFASs to aquatic systems (Müller et al., 2011).

#### 3.3.1. Well depth

Well depth was negatively correlated with all three metrics of OWC occurrence (Table 3). Compared to wells  $\geq 15$  m in depth, wells  $\leq 15$  m had higher median  $N_{\text{detects}}$  (8 vs. 4 compounds),  $\Sigma[\text{pharma}]$  (5.0 vs. 0.4 ng/L) and  $\Sigma[\text{PFAS}]$  (6.0 vs. 0.5 ng/L). In general, shallower wells pull younger groundwater more recently in contact with the atmosphere (Plummer and Friedman, 1999) with shorter flow paths that allow less time for sorption and biodegradation processes. Shallower wells have previously been found to show the greatest impact from

septic systems and other pollution sources. Among over 3800 domestic wells in Wisconsin, USA, deeper wells tended to have lower  $[\text{NO}_3^-]$  (Knobeloch et al., 2013). In 26 shallow domestic wells in a sand and gravel aquifer, shallower wells ( $\leq 8$  m) had higher concentrations of pharmaceuticals and were more likely to have reducing conditions (Verstraeten et al., 2005). In a survey of OWCs in 47 US wells suspected to be impacted by septic systems, landfills, animal feedlots, and other contamination sources (Barnes et al., 2008), well depth was negatively correlated with  $N_{\text{detects}}$ , and the median  $N_{\text{detects}}$  was more than twice as high in shallow wells ( $\leq 10$  m) compared to deeper wells ( $> 50$  m).

Shallower wells do not always contain more OWCs than deeper wells. Fram and Belitz (2011) did not find a difference in well depth

Table 3

Spearman  $\rho$  correlation coefficients between factors associated with septic system impact and three metrics of OWC presence: total number of detected OWCs ( $N_{\text{detects}}$ ), sum of detected pharmaceutical concentrations ( $\Sigma[\text{pharma}]$ ) and sum of detected PFAS concentrations ( $\Sigma[\text{PFAS}]$ ). RES = average density of residential development.

	[Nitrate]	[Total N]	[Boron]	[Acesulfame]	RES	Well depth
$N_{\text{detects}}$	0.59 <sup>***</sup>	0.59 <sup>***</sup>	0.81 <sup>***</sup>	0.87 <sup>***</sup>	0.48 <sup>*</sup>	−0.68 <sup>***</sup>
$\Sigma[\text{pharma}]$	0.34	0.34	0.59 <sup>***</sup>	0.69 <sup>***</sup>	0.43 <sup>*</sup>	−0.51 <sup>***</sup>
$\Sigma[\text{PFAS}]$	0.48 <sup>*</sup>	0.48 <sup>*</sup>	0.75 <sup>***</sup>	0.83 <sup>***</sup>	0.34	−0.64 <sup>***</sup>

\*\*\*  $p \leq 0.001$ .

\*\*  $p \leq 0.01$ .

\*  $p \leq 0.05$ .

<sup>\*</sup>  $p \leq 0.1$ .

between wells with detectable concentrations of pharmaceuticals and wells with no detectable pharmaceuticals in a study of 1200 wells in California. The California wells were much deeper than the wells we tested on Cape Cod (median depth: 134 m versus 18 m). Differences in well depth may be less important in deeper wells, since most attenuation of OWCs occurs in upper soil layers. Differences in aquifer conditions are also related to depth. Among 29 domestic wells in Minnesota, USA, deeper bedrock wells (33–87 m) tended to have higher concentrations of sulfamethoxazole and carbamazepine than shallower wells (9–34 m) in a glacial sand and gravel aquifer (Erickson et al., 2014). Higher dissolved oxygen and lower specific conductance in the deeper wells indicated that the water pulled from the bedrock wells had shorter duration flow paths. Thus, while some studies have found inverse associations between well depth and OWC detections, well depth may not be a good predictor of OWCs in deeper well systems and when comparing multiple aquifer types.

### 3.3.2. Land use

The average density of residential development in well capture zones (described in Section 2.1) was not consistently correlated with OWC presence. Only one of the three metrics of OWC presence,  $N_{\text{detects}}$ , was significantly correlated with average density (Table 3). Nevertheless, the correlation between  $N_{\text{detects}}$  and average density indicates that our approach is a useful screening tool for OWCs in domestic wells, especially in the absence of chemical testing data.

The lack of stronger correlations between average density and OWCs may be due in part to our limited ability to accurately predict well capture zones. There is considerable uncertainty associated with defining capture zones for domestic wells, especially in the absence of site-specific hydrogeological data (Cole and Silliman, 1997). The size and location of capture zones are influenced by pumping volume, well depth, hydraulic conductivity, and direction of groundwater flow. Heterogeneity in hydraulic conductivity can lead to small-scale variations in groundwater velocity and direction of groundwater flow (Cole and Silliman, 1997; Francke et al., 1998). Direction of groundwater flow can vary seasonally and can be influenced by pumping withdrawals from an aquifer.

The method we used to develop capture zones was based on typical groundwater conditions on Cape Cod and accounted for fluctuations in flow paths up to 30° (Kerfoot and Horsley, 1988). However, our approach did not take into account the location of each well relative to the dominant direction of groundwater flow, which is an important determinant of capture zone location. The Cape Cod aquifer has six groundwater lenses, with their maximum elevations along the midpoint of the peninsula. For wells near the center of each lens, capture zones are likely to be relatively close, since groundwater primarily flows in a vertical direction (Masterson and Walter, 2009), whereas for wells near the edges of each lens, capture zones are likely to be more distant, since the groundwater flows in a primarily horizontal direction.

Other approaches have been developed to assess land use near drinking water wells in the absence of site-specific hydrogeological data. In public supply wells, 500-m radius circular zones have been used to relate land use to concentrations of pollutants, such as volatile organic compounds (Johnson and Belitz, 2009). In our study of Cape Cod public supply wells (Schaider et al., 2014), we found that the extent of unsewered residential land use within 500 m of public supply wells on Cape Cod was significantly correlated with  $N_{\text{detects}}$  and  $\Sigma[\text{pharma}]$ . However, capture zones for domestic wells are smaller due to their smaller pumping volumes, and because passive wells such as domestic wells tend not to draw down the water table (Cole and Silliman, 1997), they are expected to extend primarily in an upgradient direction. In addition, since capture zones for domestic wells are relatively small, they receive inputs from a limited number of individual homes, so chemical concentrations may be more variable depending on the characteristics of the wastewater from those individual homes.

The presence of OWCs in most of the wells we tested suggests that current regulations specifying minimum distances between septic systems and domestic wells (in the US, typically 30 m; Wilcox et al., 2010) are not adequate to protect domestic wells from contamination by septic systems. Groundwater modeling suggests that in areas with one septic system per 2000–20,000 m<sup>2</sup> (0.5–5 acre lots), at least 40% of domestic wells will pump water that comes in part from septic systems (Bremer and Harter, 2012). On Cape Cod, 50% of the wells we tested had “medium density” residential development in their capture zones (0.25–0.5 acre lots), equivalent to one system per 1000–2000 m<sup>2</sup>, suggesting that more than 40% of wells pump water from septic systems. Only two of the 20 wells we tested had “very low density” development with N1 acre lots (N4000 m<sup>2</sup>) in their capture zones.

### 3.3.3. Nitrogen, boron, and TOC

All three metrics of OWC presence were more strongly correlated with [B] than with  $[\text{NO}_3^-]$  (Table 3). The three wells without detectable OWCs all contained background [B] (b 10 µg/L). Wells with N20 µg/L B had 3.8 times more detectable OWCs, 7.4 times higher  $\Sigma[\text{PFAS}]$ , and 8.8 times higher  $\Sigma[\text{pharma}]$  than wells with 10–20 µg/L B (Fig. 3). Some wells, particularly those in close proximity to the coastline, may have derived some of their B from seawater intrusion or coastal flooding ([B] in seawater: ~4.4 mg/L); concentrations of B and sodium (Na) were correlated ( $p = 0.68$ ,  $p < 0.001$ ). However, the two wells with the highest sodium concentrations (44, 45 mg/L) were not the wells with the highest [B], and the strong correlation between [B] and OWCs suggests that saltwater was the primary major source of B to the wells tested in this study.

Compared to wells with b 1 mg/L  $\text{NO}_3^-$ , wells with N1 mg/L  $\text{NO}_3^-$  had three times as many detectable OWCs, 6.2 times higher  $\Sigma[\text{PFAS}]$  and higher  $\Sigma[\text{pharma}]$  (2 vs. 0 ng/L). While 1 mg/L  $\text{NO}_3^-$  is just one-tenth of the US drinking water standard, our findings suggest significant presence of OWCs even in water with  $[\text{NO}_3^-]$  well below the drinking water standard. Nevertheless, the weaker correlations between OWC presence and  $[\text{NO}_3^-]$  suggests that while septic systems are the largest source of  $\text{NO}_3^-$  inputs into Cape Cod groundwater, there may be additional sources of  $\text{NO}_3^-$  to groundwater in the vicinity of some wells. While [B] and  $[\text{NO}_3^-]$  were strongly correlated (Spearman  $p = 0.7$ ,  $p < 0.001$ ), the combination of both [B] and  $[\text{NO}_3^-]$  can help distinguish wells that receive  $\text{NO}_3^-$  or B from other sources. For instance, the well with second highest  $[\text{NO}_3^-]$  (8 mg/L) had 21 µg/L [B] (just below the median) and ~0.4 ng/L acesulfame (lowest detected concentration), and no other detected OWCs, suggesting that the relatively high  $[\text{NO}_3^-]$  came from another source. In wells with lower  $[\text{NO}_3^-]$  than expected based on [B], it is possible that some total nitrogen was lost through denitrification or assimilation into organic N. In some shallow groundwater systems close to septic system discharges where reducing conditions predominate, the majority of TN can be present as  $\text{NH}_4^+$  (Verstraeten et al., 2005; Swartz et al., 2006). Additional analyses of N and B isotopic signatures could further distinguish domestic wastewater from other potential sources (Vengosh et al., 1994).

Wells with TOC N 1 mg/L had higher median  $N_{\text{detects}}$  (10 vs. 5 compounds),  $\Sigma[\text{pharma}]$  (7 vs. 0.4 ng/L), and  $\Sigma[\text{PFAS}]$  (11 vs. 0.8 ng/L) compared to wells with TOC b 1 mg/L. We were unable to test for correlations between TOC and metrics of OWC concentrations because TOC was only detectable in five wells.

### 3.3.4. Acesulfame

Acesulfame was the most frequently detected OWC and was always present in samples that contained detectable concentrations of any other OWC. Acesulfame concentrations were more strongly correlated with  $N_{\text{detects}}$ ,  $\Sigma[\text{pharma}]$ , and  $\Sigma[\text{PFAS}]$  than were concentrations of B and  $\text{NO}_3^-$ . Acesulfame concentrations have previously been correlated with concentrations of: PFASs in Swiss rivers (Müller et al., 2011); three pharmaceuticals (carbamazepine, gemfibrozil, naproxen) in a



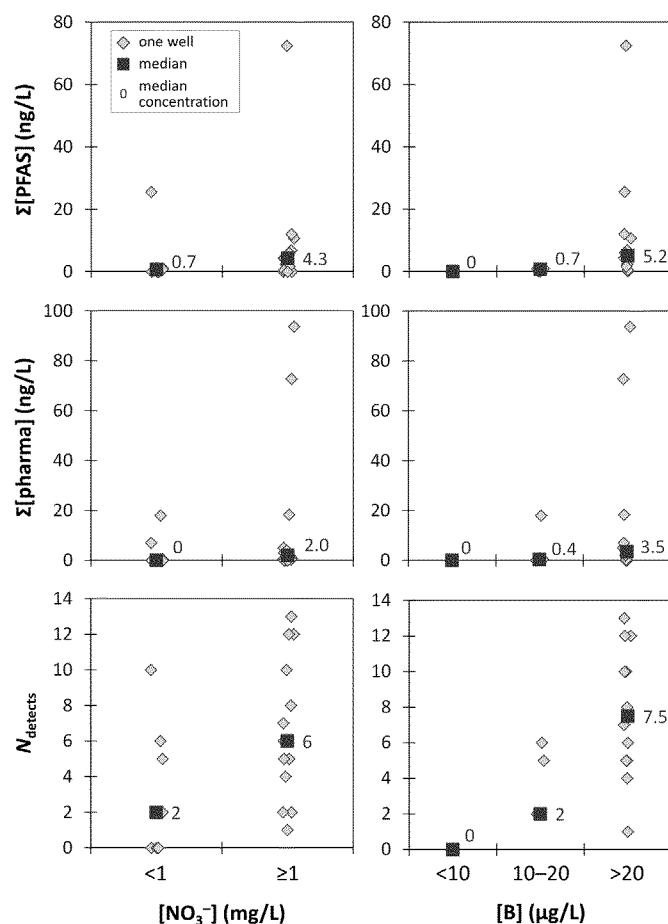


Fig. 3. Three metrics of OWC presence in 20 domestic wells, categorized according to nitrate and boron concentrations.

wastewater-impacted river in Canada (Liu et al., 2014); two pharmaceuticals (carbamazepine, sulfamethoxazole) in groundwater impacted by septic systems; and two pharmaceuticals (carbamazepine, primidone) and a nicotine metabolite (cotinine) in groundwater impacted by a municipal wastewater plume (Van Stempvoort et al., 2013).

Acesulfame has been considered an ideal marker of wastewater due to its relatively high concentrations in domestic wastewater, lack of industrial or agricultural sources, and resistance to biodegradation and sorption (Buerge et al., 2009). Acesulfame is commonly used in beverages, food, and toothpastes (Buerge et al., 2009) and can enter domestic wastewater through excretion and direct product disposal (e.g., pouring out unconsumed beverages). It is frequently detected in surface and groundwater impacted by wastewater. Acesulfame was persistent for at least 15 years in a septic system plume, with concentrations of 8000–18,000 ng/L 200 m from the leach field (Robertson et al., 2013). Acesulfame and another artificial sweetener, sucralose, underwent negligible removal during wastewater treatment and groundwater transport, whereas two other artificial sweeteners, cyclamate and saccharin, showed  $\geq 90\%$  removal in wastewater treatment plants (Buerge et al., 2009), in a septic system plume (Robertson et al., 2013), and during riverbank filtration and artificial groundwater recharge (Scheurer et al., 2010). Among these four artificial sweeteners, acesulfame was detected at the highest concentrations in Swiss groundwater and surface waters (Buerge et al., 2009). In short, acesulfame is a sensitive marker of septic system impact and can be used as an indicator compound to identify wells that are likely impacted by other OWCs from domestic wastewater.

### 3.4. Other sources of OWCs

Septic systems and other onsite wastewater treatment systems are the major source of nitrogen to Cape Cod groundwater and are the likely source for many of the OWCs that we detected in domestic wells. All of the sampled wells are located in areas served exclusively by onsite wastewater treatment systems; these systems can discharge OWCs into groundwater. In addition to domestic wastewater, sources of OWCs into groundwater include landfill leachate, stormwater runoff, and wastewater from commercial and industrial sources (Erickson et al., 2014).

Overall,  $\Sigma[\text{PFAS}]$  was correlated with  $[\text{NO}_3^-]$ , and wells with less than 15 ng/L total PFAS generally showed a consistent relationship with  $[\text{NO}_3^-]$  (Fig. S1). This relationship is consistent with septic system leachate as the main source of PFASs for most of these wells. However, the two wells with the highest  $\Sigma[\text{PFAS}]$  (72, 26 ng/L) visibly departed from this relationship, suggesting contributions from other sources. One of these two wells, W09, was located 3.2 km from a capped landfill, although the well was not directly downgradient of the landfill. This well had the highest acesulfame concentration (5300 ng/L) and the highest concentration of three PFASs (PFHxS, PFBS, PFHxA). However, unlike most other wells with N400 ng/L acesulfame, the total detectable concentration of pharmaceuticals and personal care products (PPCPs) was relatively low (4 ng/L) in this well. Furthermore, the PFHxS concentration in well W09 (47 ng/L) was higher than the maximum concentrations reported in effluent from 23 US wastewater treatment plants (24 ng/L; Schultz et al., 2006a; Schultz et al., 2006b; Sinclair and Kannan, 2006; Loganathan et al., 2007; Plumlee et al., 2008) and four septic systems (2.5 ng/L; Subedi et al., 2015). This profile suggests that another source, possibly the landfill, was the primary source of PFASs to this well. Another well, W07, was 0.3 km from a capped landfill, and its OWC profile was similarly dominated by PFASs, with  $\Sigma[\text{PFAS}]$  of 4.3 ng/L and no detected PPCPs despite an acesulfame concentration of 1100 ng/L.

In addition to landfill and septic system leachate, there may be other sources of PFASs to domestic wells on Cape Cod. Another well (W03) with elevated  $\Sigma[\text{PFAS}]$  (26 ng/L) and relatively low  $[\text{NO}_3^-]$  (0.8 mg/L) did not appear to be located downgradient of a landfill. This well was located near an unsewered commercial area; wastewater from local businesses that use PFASs in paints, textiles coatings, or floor waxes may have been a source of PFASs to this well.

The results from our wells show the complex interactions among multiple sources of OWCs and the limitations of attributing pollution to specific sources based on a single chemical indicator. Landfill leachate contains elevated concentrations of OWCs originating from consumer products in municipal solid waste and from sewage sludge. Pharmaceuticals, personal care product ingredients, OPFRs, and PFASs have all been found in landfill leachates, with concentrations often varying by several orders of magnitude across locations (Clarke et al., 2015). PFAS concentrations in landfill leachate range from low ng/L to 100s of ng/L (Eggen et al., 2010; Huset et al., 2011; Clarke et al., 2015). These studies show that in general, perfluorocarboxylic acids (e.g., PFOA) are more abundant than perfluorosulfonic acids (e.g., PFOS) and shorter-chain PFASs are more abundant than longer-chain, although individual landfills have different relative abundances.

While acesulfame and sucralose have been considered specific markers of wastewater in aquatic systems (Buerge et al., 2009), artificial sweeteners are also common in landfill leachate, with concentrations up to 85  $\mu\text{g/L}$  acesulfame (Roy et al., 2014) and 620  $\mu\text{g/L}$  sucralose (Clarke et al., 2015). Combining information about when specific artificial sweeteners were introduced or withdrawn and the years of operation of a landfill can help distinguish multiple sources of OWCs in groundwater and can also be used to age-date landfill plumes (Van Stempvoort et al., 2011; Roy et al., 2014). Concentrations of B, and to a lesser extent nitrogen, can also be elevated in landfill leachate. The median [B] in a study of landfill leachate was 4700  $\mu\text{g/L}$  (Clark and Piskin,

1977), which is ~20 times higher than septic tank effluent (Swartz et al., 2006), whereas the median concentration of total N ( $\text{NO}_3^- + \text{NH}_4^+$ ) was 82 mg/L (Clark and Piskin, 1977), which is only around twice typical TN concentrations in septic tank effluent (Katz et al., 2010). Thus in groundwater systems with multiple possible sources of OWCs into drinking water wells, a combination of indicators are needed to distinguish multiple sources.

### 3.5. Implications

The results of our domestic well testing on Cape Cod demonstrate that domestic well water quality is affected by OWCs from septic systems and other sources. Our finding that OWC presence was correlated with concentrations of  $\text{NO}_3^-$  and B is useful since it can be difficult to predict which wells are likely to contain OWCs from land use data alone due to uncertainties in determining domestic well capture zones. Detailed hydrogeological models that account for small-scale variations in hydraulic conductivity and flow direction can be used to model flow paths for water pumped from domestic wells, but these detailed models often are not available to planners and developers making decisions about siting domestic wells (Wilcox et al., 2010). Regulations provide minimum well-casing depths and minimum setback distances between wells and septic system tanks and leach fields (in the US, typically 15 m from septic tanks and 30 m from leach fields; Wilcox et al., 2010). While these regulations are designed to protect domestic wells from wastewater pathogens, waterborne disease outbreaks can still occur from drinking water wells contaminated by new septic systems built in compliance with local regulations (Borchardt et al., 2011). Epidemiological studies have shown associations between septic system density and diarrheal disease in children (Borchardt et al., 2003) and have found septic systems to be a common contributing factor to disease outbreaks associated with untreated groundwater in the US (Wallender et al., 2014). To protect water quality in new residential developments, Wilcox et al. (2010) suggests that installing a single larger volume well in an area upgradient of the development, rather than individual wells on each property, may offer better water quality protection in areas that are not served by public water supplies.

Even when pathogens from septic systems are adequately treated to prevent waterborne disease, septic systems are also sources of OWCs to domestic wells. The OWCs we detected in Cape Cod domestic wells are not currently regulated by enforceable drinking water standards and health-based guideline values are only available for 10 of the 27 OWCs we detected. We have previously described the limitations of current efforts to assess health effects of pharmaceuticals and other OWCs in drinking water (Schaider et al., 2014). Some risk assessments have suggested that few or no health risks are associated with commonly reported drinking water exposures from pharmaceuticals, since such exposures are generally far below therapeutic levels or levels where adverse health effects have been reported in animal or human studies (Bruce et al., 2010; WHO, 2011). However, few guidelines for pharmaceuticals have been developed on the basis of toxicity studies, and toxicity assessment for pharmaceuticals may not include rigorous assessment of developmental toxicity, neuro- or immunotoxicity, endocrine toxicity, and carcinogenicity (ICH, 2013).

Among the chemicals we detected, PFASs may be of greatest health concern given their relatively high detection frequencies and evidence of health effects at low doses. Maximum concentrations of PFOS and PFOA in this study were well below the US EPA's provisional health guidelines (200 and 400 ng/L, respectively). However, these guidelines were developed for short-term exposures; the state of New Jersey has set a PFOA guideline for drinking water that is ten times lower (Post et al., 2009). Furthermore, epidemiological studies suggest that current guideline values do not sufficiently protect children from immunotoxic effects (Grandjean and Budtz-Jørgensen, 2013). While little toxicological information is available for other PFASs, there is growing recognition

that unidentified PFAS precursors in contaminated groundwater can undergo transformation to form PFOS and PFOA (Houtz et al., 2013).

Our results suggest that current regulations to protect domestic wells from pathogens in septic system discharges do not prevent OWCs from reaching domestic wells. In the absence of adequate toxicological data and drinking water standards to assess the potential health effects of individual OWCs in drinking water, nitrate and boron can be used to identify drinking water sources likely to contain the highest levels of OWCs. Elevated nitrate levels can indicate wastewater impact in aerobic conditions, while under anaerobic conditions other indicators, such as total nitrogen, boron, and artificial sweeteners, are more appropriate. We found that nitrate concentrations of 1 mg/L  $\text{NO}_3^-$ -N, which are tenfold higher than local background and tenfold lower than the US federal drinking water standard, were associated with wastewater impacts from OWCs. Since nitrate is a commonly measured drinking water contaminant, it is a useful screening tool for OWCs in domestic wells and target nitrate levels can be established to prioritize wells for additional water quality testing and enhanced source water protection.

### 4. Conclusions

In this study, we found organic wastewater compounds, including per- and polyfluoroalkyl substances, pharmaceuticals, and organophosphate flame retardants, in shallow domestic drinking water wells in a sand and gravel aquifer where septic systems are prevalent. Concentrations of some pharmaceuticals and PFASs were higher than other US drinking water sources. This is the first study to show PFASs in domestic wells that are not impacted by production facilities, aqueous film forming foams, or aviation sources. The presence of OWCs in drinking water raises human health concerns, but a full evaluation of potential risks is limited by a lack of health-based guidelines and toxicological assessments. The presence of OWCs was correlated with concentrations of nitrate, boron, and acesulfame (artificial sweetener), and inversely correlated with well depth. In addition to septic systems, landfill leachate and wastewater from commercial development also may be sources of PFASs and other OWCs. Our results demonstrate the likelihood of OWC contamination in drinking water supplies in sand and gravel aquifers where groundwater discharges of wastewater are prevalent, especially in wells with elevated concentrations of nitrate and boron. Nitrate testing is an inexpensive marker of OWCs in aerobic conditions and is readily available for domestic well owners and local boards of health, who can use the results to identify vulnerable wells for additional testing and enhanced source water protection.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.scitotenv.2015.12.081>.

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